



Rec'd PCT/PTO 20 JUN 2005
PCT 2003 / 0 0 5 5 2 1



INVESTOR IN PEOPLE

#2

**PRIORITY
DOCUMENT**

SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH RULE 17.1(a) OR (b)

The Patent Office
Concept House
Cardiff Road
Newport
South Wales
NP10 8QQ

REC'D 20 JAN 2004

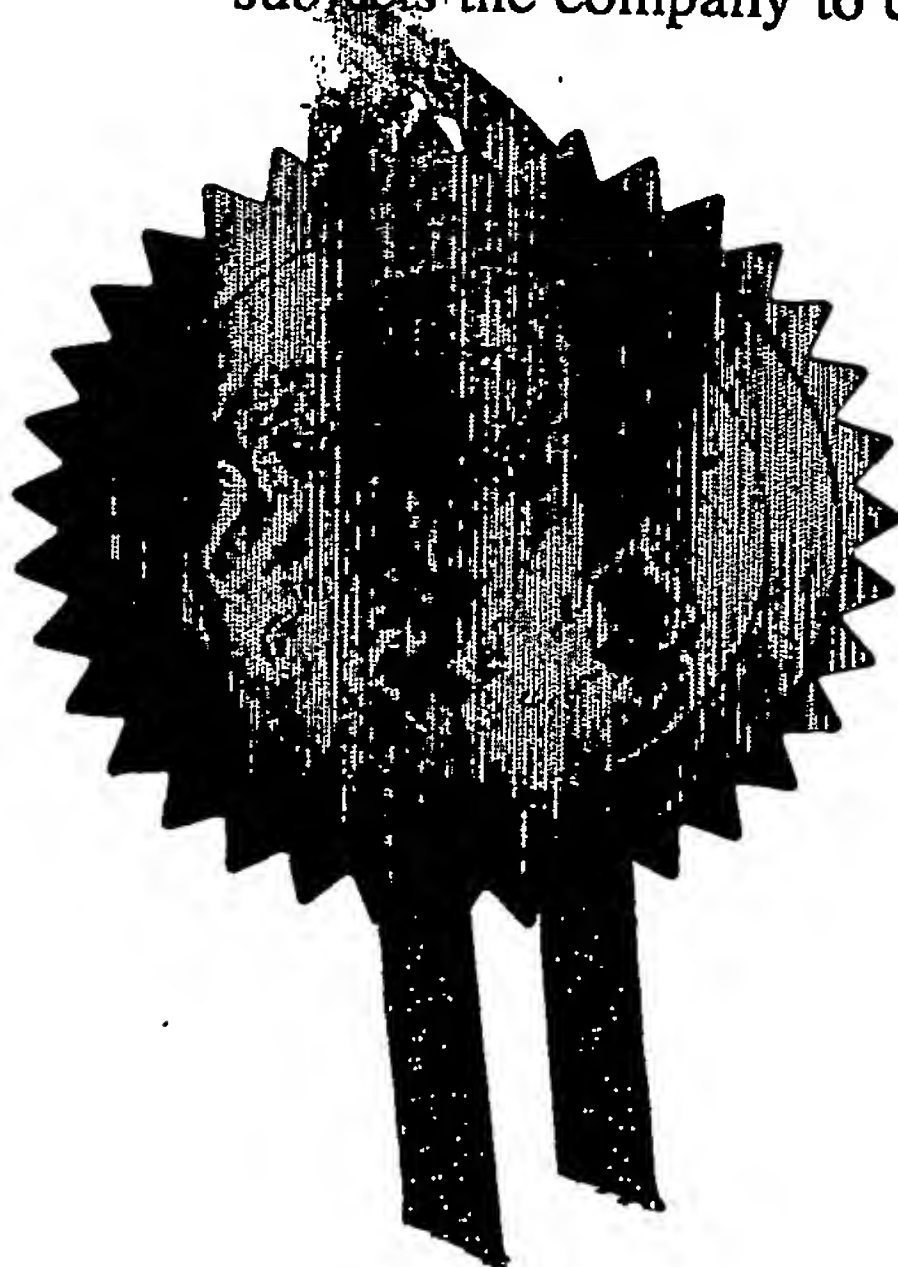
WIPO PCT

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

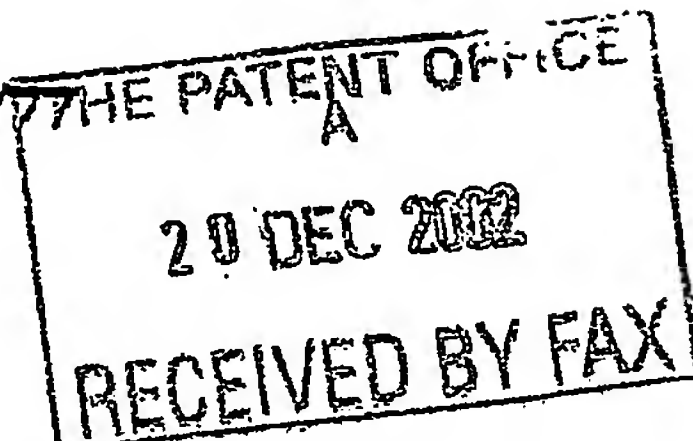


Signed

Dated 12 January 2004

BEST AVAILABLE COPY

Patents Form 1/77

The
Patent
OfficePatents Act 1977
(Rule 16)20DEC02 E772525-2 P01/77
P01/77 0.00-0229660.6**Request for grant of a patent**

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form.)

The Patent Office

Cardiff Road
Newport
Gwent NP9 1RH

1. Your reference

SMC 60569/GB/P1

2. Patent application number

(The Patent Office will fill in this part)

0229660.6

20 DEC 2002

3. Full name, address and postcode of the or of each applicant (underline all surnames)

Avecia Limited
Hexagon House
Blackley
Manchester, M9 8ZS

Patents ADP number (if you know it)

07764137001

If the applicant is a corporate body, give the country/state of its incorporation

GB

4. Title of the invention

ELECTRONIC DEVICES

5. Name of your agent (if you have one)

PARLETT, Peter Michael

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

Avecia Limited
Hexagon House
Blackley
Manchester, M9 8ZS
United Kingdom

Patents ADP number (if you know it)

8463655001

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number
(if you know it)Date of filing
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if)

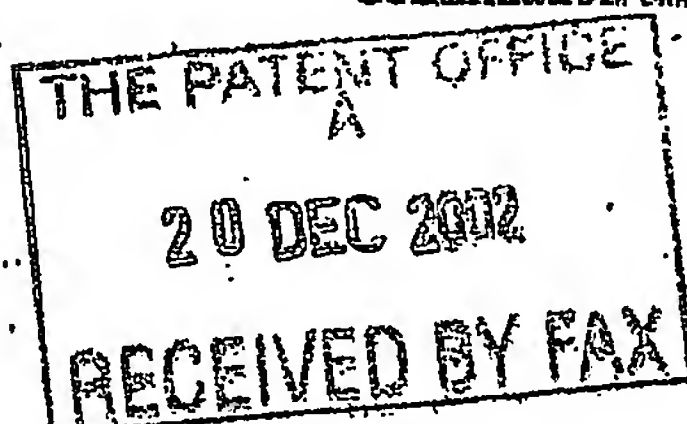
- a) any applicant named in part 3 is not an inventor, or
b) there is an inventor who is not named as an applicant, or
c) any named applicant is a corporate body.
See note (d))

Yes

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document

Continuation sheets of this form



Description

21 ✓

Claim(s)

09 ✓

Abstract

04 ✓

Drawing(s)

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

01 ✓

Request for substantive examination (Patents Form 10/77)

Any other documents
(please specify)

11.

I/We request the grant of a patent on the basis of this application.

Signature

K.M. Pinder

Date

20/12/02

Avecia Limited Authorised Signatory

12. Name and daytime telephone number of person to contact in the United Kingdom

Mrs K.M. Pinder/Miss G. Terry 0161 721 1361/2

Warning

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

Notes

- If you need help to fill in this form or you have any questions, please contact the Patent Office on 0645 500505.
- Write your answers in capital letters using black ink or you may type them.
- If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.
- If you have answered 'Yes' Patents Form 7/77 will need to be filed.
- Once you have filled in the form you must remember to sign and date it.
- For details of the fee and ways to pay please contact the Patent Office.

SMC 60569

1

Cathode
EIL
ETL1
ETL2
EL layer
HTL2
HTL1
HIL (e.g. PAni or PEDOT)
Anode(e.g. ITO)

Figure 1 (Prior art)

NOT TO BE AMENDED

SMC 60569

2

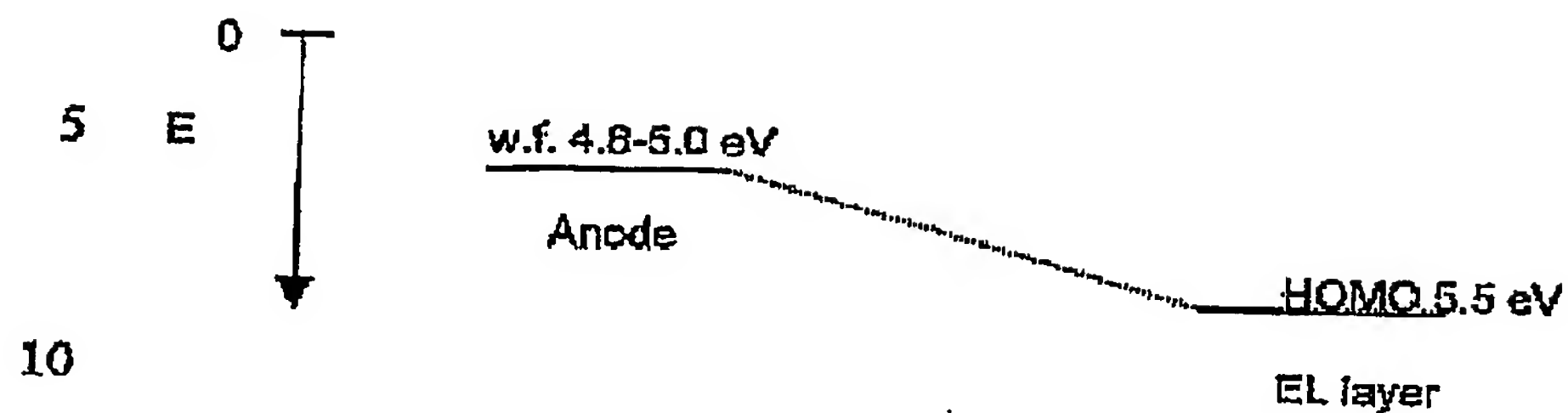
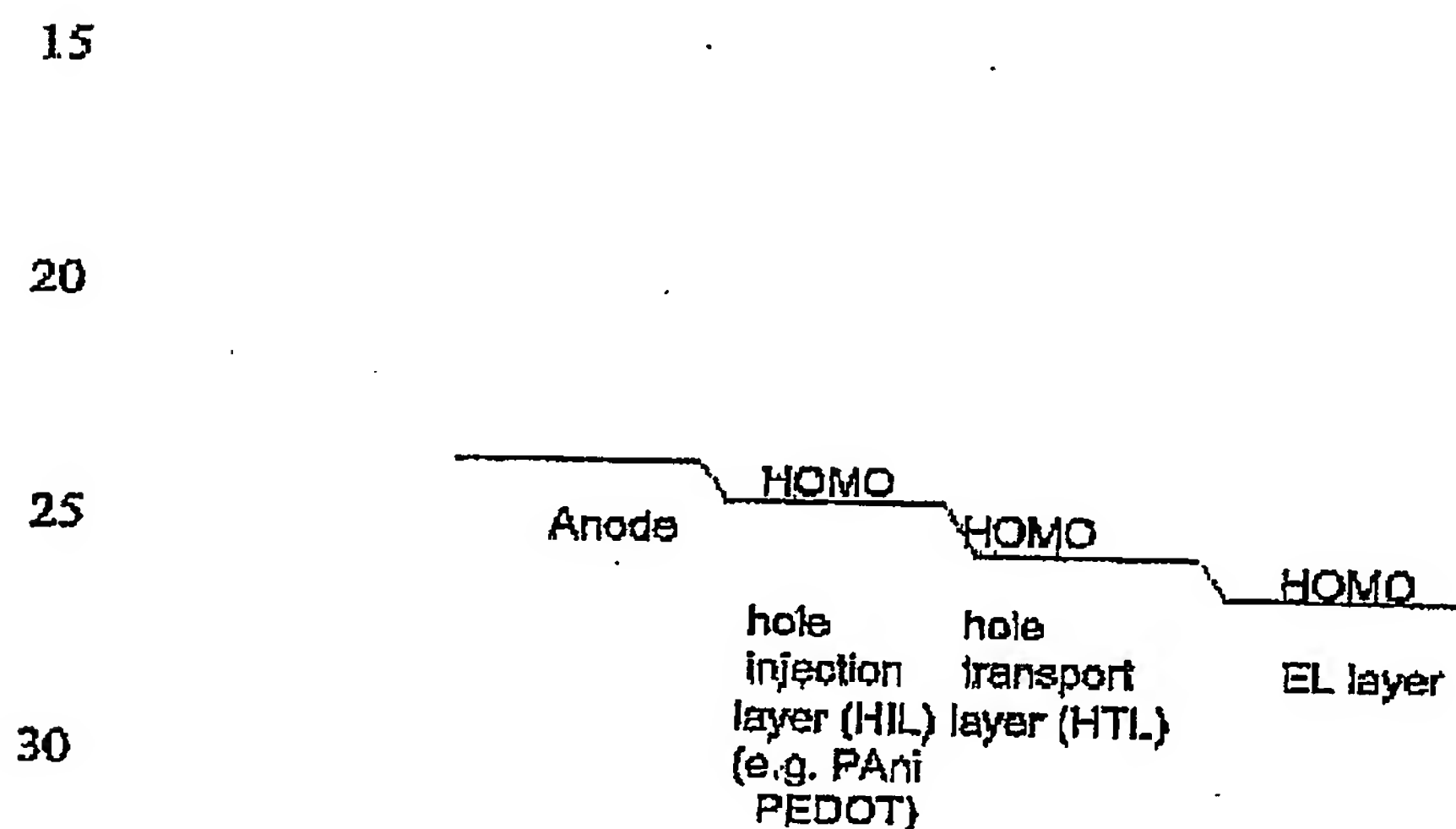


Figure 2A (Prior art)



35 Figure 2B (Prior art)

NOT TO BE AMENDED

SMC 60569

3

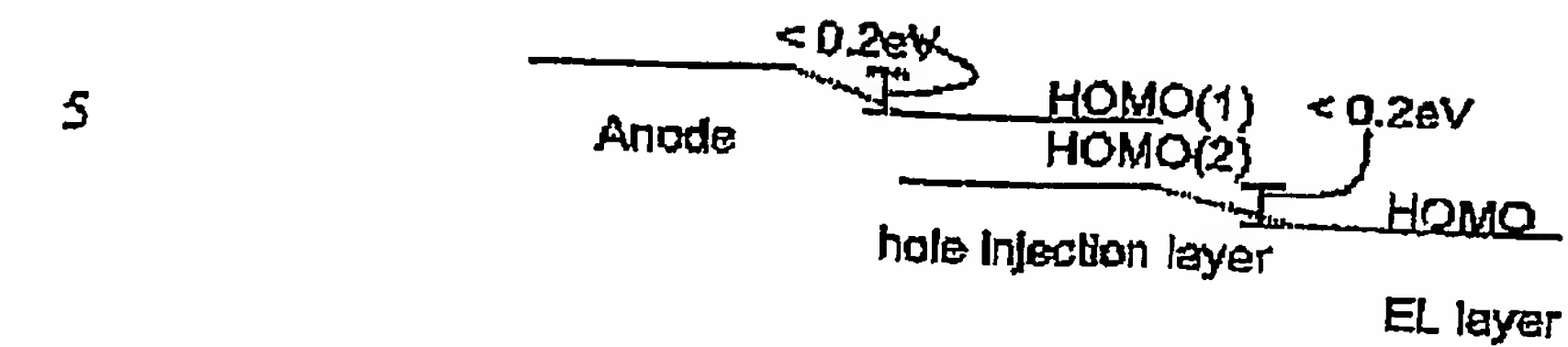


Figure 3 (Invention)

15

NOT TO BE AMENDED

SMC 60569

4

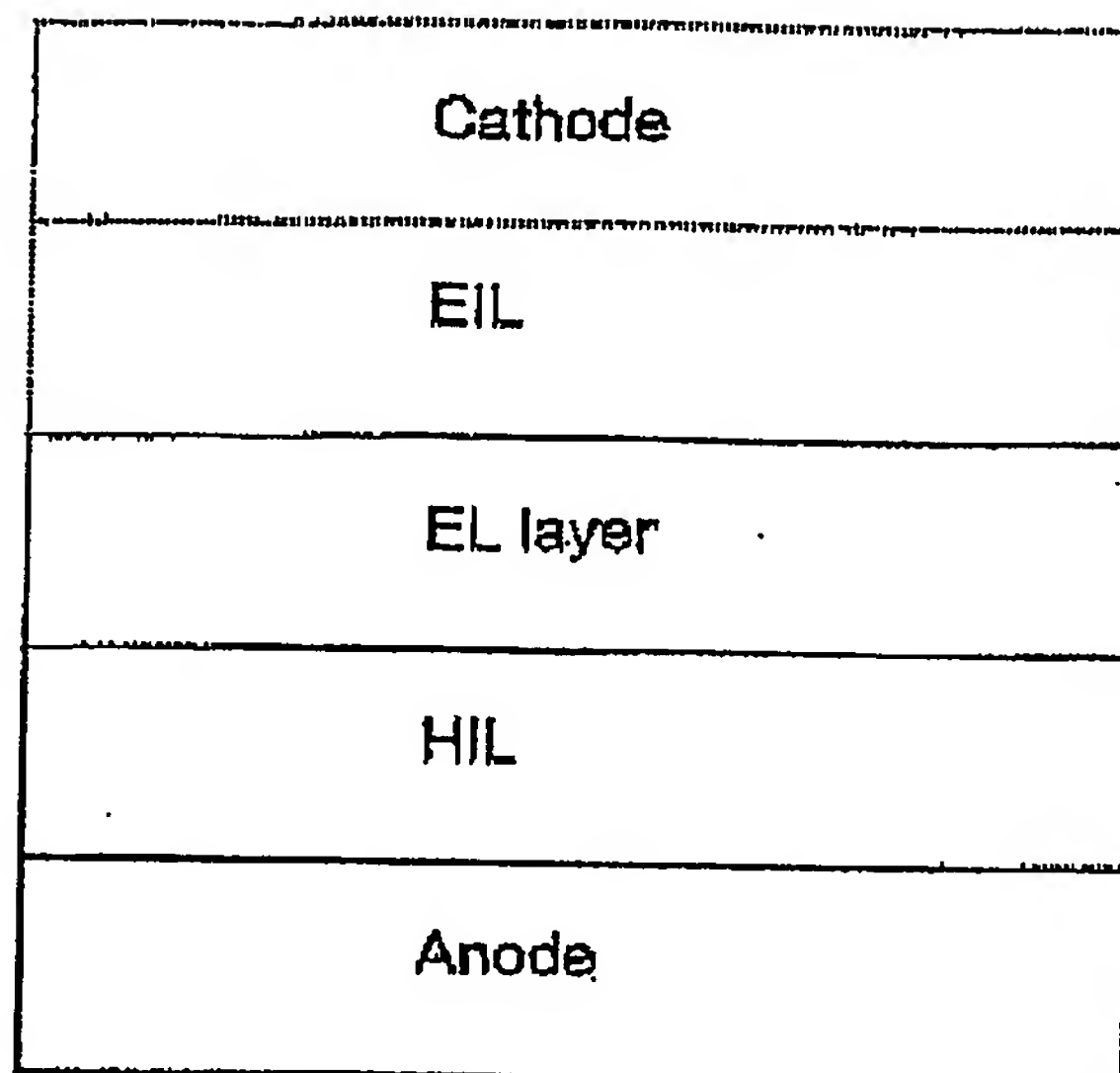


Figure 4
(Invention)

NOT TO BE REPRODUCED

20-DEC-2002 16:59 FROM: AVECIA LTD

01617215801

TO: +01 814444

P.005

SMC 60569

DUPLICATE

APPLICANTS

AVECIA LIMITED

TITLE

ELECTRONIC DEVICES

SMC 60569

1

DUPLICATE

ELECTRONIC DEVICES**Field of the Invention**

5 The present invention concerns improvements in and relating to electroluminescent (EL) devices such as Organic Light Emitting Diodes (OLEDs).

Background of the Invention

10 OLEDs are optoelectronic devices being developed for use in flat panel displays as alternatives to existing technologies such as the cathode ray tube and liquid crystal displays. OLEDs have the potential to offer numerous advantages including being lightweight and non-bulky, low powered, wide viewing angled, applicable to large display areas and cheaper to manufacture.

15 An OLED device comprises an organic electroluminescent (EL) layer located between two electrodes. At least one of the electrodes is transparent to allow transmission of light from the EL layer. In operation, when a voltage is applied across the device via the electrodes, holes are injected into one side of the EL layer from one electrode (the anode) and electrons are injected into the other side from the other electrode (the cathode). The holes and electrons move through the EL layer in opposite directions under the applied voltage until they meet and recombine to form an excited state which subsequently decays with the emission of light.

20 Holes should effectively transfer from the anode into the highest occupied molecular orbital (HOMO) energy level of the EL layer. Similarly, electrons should effectively transfer from the cathode into the lowest unoccupied molecular orbital (LUMO) energy level of the EL layer. Unfortunately, the workfunction of the anode often differs from the HOMO level of the EL layer. The same problem also exists between the cathode and the LUMO level of the EL layer. In practice, since the relevant electronic energy levels of the anode and cathode are often not ideally matched to the HOMO and LUMO respectively of the organic EL material, it is usual to modify the basic OLED structure to improve energy level matching and thus improve hole and/or electron injection efficiency. A extensively employed method is to use between the EL layer and the anode and/or 30 cathode respectively an additional organic layer. These additional layers are commonly referred to as a hole injection layer (HIL) and an electron injection layer (EIL). The HIL and/or EIL improve matching of the energy levels, thus improving efficiency of the device. In addition, the HIL and EIL may serve other purposes such as electron blocking and hole blocking respectively (i.e. the HIL or EIL are each one type /one direction carrier transport materials). It has become typical to employ as HIL on the anode a layer of a conducting 35 organic material, such as polyaniline (PAni) or polyethylenedioxythiophene (PEDOT).

40 It is not uncommon to find devices employing one or more further organic layers between the HIL and/or EIL respectively and the EL layer to achieve even further enhanced matching of energy levels and thus still higher operating efficiency. The further organic layers between the HIL or EIL and the EL layer are termed hole transport layers (HTLs) and electron transport layers (ETLs) respectively to differentiate them from the HIL

SMC 60569

2

or EIL which directly contact the anode or cathode respectively. Such terminology is used herein. A prior art device structure is shown schematically in Figure 1.

Despite the above developments in EL devices which has occurred over recent years, in many aspects there still remains a need for improvement. The present invention is concerned with improvements relating to organic layers in EL devices particularly, but not exclusively, in HILs and HTLs, as well as in EILs and ETLs.

In connection with HILs/HTLs and EILs/ETLs, examples of areas where performance could be improved include efficiency and drive voltage, transparency, coatability, chemical stability and lifetime. Moreover, present device structures consisting of multiple layers for charge carrier injection/transport make manufacture complex and costly. It would be desirable to achieve similar or improved performance using a single layer in place of the present multiple layers. In addition, in a colour display, EL layers of different colours have different HOMO/LUMO levels making it difficult to use the same charge injection/transport layers for pixels of each colour without loss of performance.

Objects of the invention include reducing or overcoming one or more of the above problems of the prior art. Further objects of the invention will be apparent from the following description.

Statement of the Invention

According to the present invention there is provided an electroluminescent device having an anode, a cathode and one or more organic layers between said anode and said cathode, at least one of said organic layers comprising an organic electroluminescent material, wherein at least one of said organic layers comprises a composition having at least two HOMO energy levels and/or at least two LUMO energy levels.

In use, when a voltage is applied between the anode and the cathode, holes are injected into the layer adjacent the anode and electrons are injected into the layer adjacent the cathode, whereby light is emitted from the at least one layer comprising the electroluminescent material.

Advantageously, the use of the composition according to the invention reduces the number of charge injection/transport layers required whilst providing good performance as will be described in more detail below.

Detailed Description of the Invention

The EL device is preferably an OLED. At least one of the anode or cathode is transparent.

The device may comprise only one organic layer in which case the organic layer comprising the composition of the invention is the same layer as the layer comprising the EL material. Typically, the device comprises two or more organic layers, wherein the organic layer comprising the composition of the invention is different to the layer comprising the EL material. The at least one organic layer comprising the composition has

SMC 60569

3

been found to be excellent for forming a hole injection layer (HIL) and/or hole transport layer (HTL), i.e. separate from the EL layer and located between the EL layer and the anode. Alternatively or additionally, an at least one organic layer comprising the composition may form an electron injection layer (EIL) and/or electron transport layer (ETL), i.e. separate from the EL layer and located between the EL layer and the cathode.

Each of the HOMOs and/or LUMOs can be a hole transporting and/or electron transporting level respectively.

Preferably, the energy of at least one of the HOMO levels (or LUMO levels) is within ± 0.2 eV of an electronic energy level in an adjacent layer. Additionally preferably, the energy of at least another of the HOMO levels (or LUMO levels) is within ± 0.2 eV of an electronic energy level in another adjacent layer. In one embodiment, preferably, the energy of at least one of the HOMO levels (or LUMO levels) is within ± 0.2 eV of the work function of the anode (or cathode). Additionally preferably, the energy of at least one of the HOMO levels (or LUMO levels) is within ± 0.2 eV of the HOMO level (or LUMO level) of the EL layer.

Preferably, the energy difference between the at least two HOMO energy levels and/or between the at least two LUMO energy levels is greater than 0.1 eV, more preferably greater than 0.2 eV.

Preferably, the composition comprises at least two charge transport materials, the energies of the HOMOs of the materials and/or the energies of the LUMOs of the materials being different such that the composition has at least two HOMO energy levels and/or at least two LUMO energy levels.

Preferably, the at least two charge transport materials comprise at least one polymeric material and at least one non-polymeric material preferred features of which are described below. The composition may contain two or more different polymeric materials and/or two or more different non-polymeric materials, for instance when used with pixels of different colours as described in more detail below.

Preferably, the at least two charge transport materials are organic materials and more preferably are organic semiconductors. The materials may be positive charge (hole) carrier materials, herein termed "p-type" materials. In this case, the layer comprising the composition may be a hole injection layer (HIL) or hole transport layer (HTL). Alternatively, the materials may be negative charge (electron) carrier materials, herein termed "n-type" materials. In this case, the layer comprising the composition may be an electron injection layer (EIL) or electron transport layer (ETL).

Preferably, the polymeric material of the composition is an organic material. Additionally preferably, the polymeric material is a semiconductor. Preferably, the non-polymeric material of the composition is an organic material. Additionally preferably, the non-polymeric material is a semiconductor.

SMC 60569

4

The polymeric material may be a "p-type" material. The non-polymeric material may likewise be a p-type material. In this case, the layer comprising the composition may be a hole injection layer (HIL) or hole transport layer (HTL).

5 The polymeric material may be a "n-type" material. The non-polymeric material may likewise be a n-type material. In this case, the layer comprising the composition may be an electron injection layer (EIL) or electron transport layer (ETL).

10 In known devices, the HOMO energy level (which can be measured by the ionisation potential (I_p)) of the EL layer is more often than not at a different energy than the work function (w.f.) of the anode (usually the HOMO of the EL layer is higher, e.g. 5.5 eV compared to a WF of 4.8- 5.0 eV for an Indium tin oxide (ITO) anode) as shown in Figure 2A. Similarly, the LUMO energy level of the EL layer is often at a different energy than the WF of the cathode. Consequently, there is a difficulty due to this energy barrier in trying to inject holes (electrons) into the EL layer from the anode (cathode). For this reason, in the prior art, one or more intermediate layers, e.g. HIL, HTL, (EIL, ETL) having intermediate HOMO (LUMO) levels are typically employed between the anode (cathode) and the EL layer in an attempt to bridge the energy gap and improve hole (electron) injection efficiency. In order to achieve the best injection efficiencies, two or more layers, e.g. both HIL and HTL (EIL and ETL) are employed as shown in Figure 2B. However, the more layers present, the more complex and costly the device becomes. The present invention reduces this problem as now described.

20 For the avoidance of doubt, whilst the invention may be illustrated in many instances herein by reference to use of HOMO levels and use as a HIL or HTL, it should be understood that the invention also applies to use of LUMO levels and use as an EIL or ETL mutatis mutandis.

25 The composition according to one embodiment of the invention has a plurality of HOMO energy levels, for example as shown in Figure 3. This is achieved, for example, by having at least two charge transport materials in the composition each having a well defined HOMO energy level. Preferably, one of the composition's HOMO levels is matched close in energy to (more preferably within ± 0.2 eV of) an energy level of one adjacent layer and another of the HOMO levels is matched close in energy to (more preferably within ± 0.2 eV of) an energy level of another adjacent layer, e.g. on the opposite side of the layer comprising the composition to the first adjacent layer. For example, one HOMO level is matched close to, preferably within ± 0.2 eV of, the w.f. of the anode positioned on one side of the layer comprising the composition and another HOMO level is matched close to, preferably within ± 0.2 eV of, the HOMO of the EL layer positioned on the other side of the layer comprising the composition as shown in Figure 3.

35 Thus, a single layer (HIL) comprising the composition may replace the function of two or more layers (HIL, HTL etc.) of the prior art, thereby making manufacture simpler. 40 The invention can improve the matching of both the anode w.f. and the HOMO of the EL

SMC 60569

5

layer using a single layer. The invention makes it possible to match commonly used anodes (and cathodes) with a wider variety of EL layers as it becomes easier to bridge the energy difference to the HOMO (or LUMO) of the EL layer. Improved energy matching to pixels of different colours (i.e. having EL layers of different colours, e.g. red, green, blue) can also be achieved by the invention using the same hole (or electron) injection layer. This can be achieved for example by using more than two charge transport materials in the composition enabling the use of the same injection layer for all colours efficiently. This could not easily be achieved previously with the same injection layer. The ability of the injection layer of the invention to supply charge carriers into an EL layer is enhanced leading to improved efficiency and reduced drive voltage.

In a preferred embodiment, the polymeric material has a HOMO level matched close in energy to, preferably within ± 0.2 eV of, the w.f. of the anode and the non-polymeric material has a HOMO level matched close in energy to, preferably within ± 0.2 eV of, the HOMO level of the EL layer. Alternatively, in another embodiment, the non-polymeric material has a HOMO level matched close to the w.f. of the anode and the polymeric material has a HOMO level matched close to the EL layer.

The HOMO energy levels of the materials, e.g. the polymeric material and the non-polymeric material, are preferably arranged such that the energy difference between them permits facile hole transfer from one material to the other within the composition thereby to allow hole transport across the layer comprising the composition as a whole.

Typically, each HOMO level has an energy distribution. The energy distribution of each individual HOMO level is typically ± 0.1 eV. The energy distribution can be maximised by factors such as increased polydispersity of the polymeric material. This energy distribution also improves energy matching both between the composition and adjacent layers and between the materials within the composition.

In many preferred embodiments the energies of at least one, preferably two, of the HOMO levels of the composition lies in the range from about 4.9 to about 5.7 eV.

As mentioned, the above features of the invention may be applied analogously to the role of electron injection layers (EIL) by making use of multiple LUMO levels. For example, in one embodiment, for an efficient EIL, one of the materials in the composition should have a LUMO level within ± 0.2 eV of the workfunction of the cathode and at least one other of the materials should have a LUMO level within ± 0.2 eV of the LUMO of the EL layer.

The at least one polymeric material and the at least one non-polymeric material may be mixed in a ratio from 10:90 to 90:10 % by weight. A mixing ratio of 40:60 - 60:40 % by weight is preferred, e.g. about 50/50 % by wt.

The number of repeat units which may be present per molecule in the polymeric material (and which can also be denoted by the integer 'n' herein) may be from 3 to 20,000, preferably 3 to 10,000, more preferably 4 to 1,000, still more preferably 5 to 500 and most preferably 6 to 100.

SMC 60569

6

The polymeric material of the invention is preferably polydisperse in order to provide a better energy distribution for the HOMO level (or LUMO level), in order to improve further the matching to the, e.g., anode or EL layer or other layer(s), as well as to the non-polymeric material within the composition. Preferably the polydispersity is from 1.1 to 5. More preferably, the polydispersity is from 1.1 to 3.

The polymeric material and/or the non-polymeric material, preferably at least the polymeric material, is preferably substituted by at least one optionally substituted long chain, linear, branched or cyclic carbyl-derived group, i.e. C_8 or longer, preferably an optionally substituted long chain, linear, branched or cyclic hydrocarbyl-derived group, most preferably alkyl or alkoxy. It has been found that with such substitution it is possible to readily form thick films, especially when solution coated, which, moreover, are stable and have a long lifetime.

The polymeric material may comprise a homopolymer or a copolymer. The term polymeric used herein includes oligomeric.

Polymeric materials that can be used in this invention include compounds and derivatives, all preferably soluble, of the following list :
 conjugated organic (including hydrocarbon) polymers such as polyarylamine (e.g. polytriarylamine), polyacene, polyphenylene, poly(phenylene vinylene), polyfluorene including oligomers of those conjugated hydrocarbon polymers; oligomeric para substituted phenylenes such as p-quaterphenyl (p-4P), p-quinquephenyl (p-5P), p-sexiphenyl (p-6P); conjugated heterocyclic polymers such as polythiophene, poly(3-substituted thiophene), poly(3,4-bisubstituted thiophene), polybenzothiophene, polyisothianaphthene, poly(N-substituted pyrrole), poly(3-substituted pyrrole), poly(3,4-bisubstituted pyrrole), polyfuran, polypyridine, poly-1,3,4-oxadiazoles, polyisothianaphthene, poly(N-substituted aniline), poly(2-substituted aniline), poly(3-substituted aniline), poly(2,3-bisubstituted aniline), polyazulene, polypyrene, pyrazoline compounds; polyselenophene; polybenzofuran; polyindole; polypyridazine; benzidine compounds; stilbene compounds; triazines; carbazole polymers ; butadiene polymers. Polymers and copolymers of the foregoing may be used. The HOMO level of these polymers and copolymers can be conveniently chosen by the chemical structure (substitution, copolymer ratio) of such polymers and adjusted between 4.9 - 5.7 eV.

A preferred polymeric material is polyarylamine, especially polytriarylamine.

A preferred class of polytriarylamine has repeat units of formula 1:

Formula 1:



SMC 60569

7

wherein:

Y^1 represents, independently if in different repeat units, N, P, S, As and/or Se, preferably N;

Ar¹ and Ar² are aromatic groups and Ar³ is present only if Y^1 is N, P, or As in which case it too is an aromatic group; wherein Ar¹ and Ar² are the same or different and represent, independently if in different repeat units, a multivalent (preferably bivalent) aromatic group (preferably mononuclear but optionally polynuclear) optionally substituted by at least one optionally substituted C₁₋₄₀ carbyl-derived groups and/or at least one other optional substituent; and Ar³ represents, independently if in different repeat units, a mono or multivalent (preferably bivalent) aromatic group (preferably mononuclear but optionally polynuclear) optionally substituted by at least one optionally substituted C₁₋₄₀ carbyl-derived group and/or at least one other optional substituent.

The arrow extending from Ar³ in Formula 1 is intended to indicate that the group may be monovalent or multivalent. If the group is monovalent the arrow denotes a bond to a suitable terminal group such as hydrogen or another substituent which is inert to coupling under the conditions of polymerisation (e.g. alkyl or aryl). If the group is multivalent (e.g. bivalent) the arrow denotes a bond to another repeat unit (i.e. the polymer chain is branched and/or cross-linked).

The polymer may have any chain terminating groups, for example, any leaving groups used in a polymerisation process by which the polymer is made, or end capping groups.

WO 99/32537 is a patent application of the applicants which describes polymers which have repeat units of Formula 1 and methods for their production. In that patent application, polymers of this type are prepared by the addition of an end capping reagent to control the molecular weight of the final polymer and hence its desirable properties as a charge transport material. Materials disclosed in that patent application may be useful as materials in the present invention. Accordingly, the contents of that patent application are incorporated herein by reference. Therefore, optionally, at least one terminal group is attached in the polymer to the Ar¹, Ar² and optionally Ar³ groups located at the end of the polymer chains, so as to cap the polymer chains and prevent further polymer growth, and at least one terminal group is derived from at least one end capping reagent used in the polymerisation to form said polymeric material to control the molecular weight thereof.

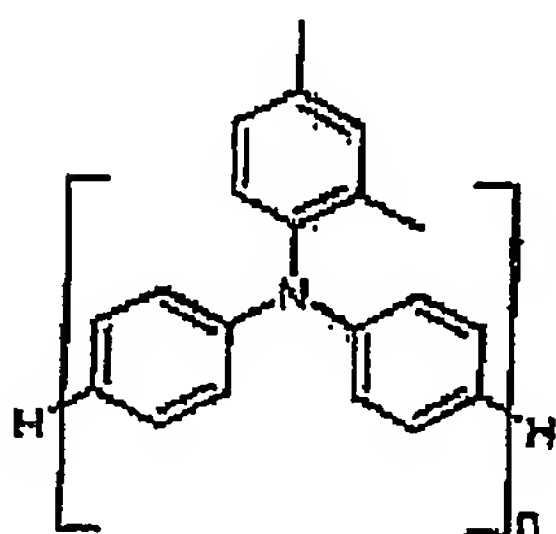
WO 00/78843 is another patent application of the applicant which describes polymers which have repeat units of Formula 1. In that patent application, the polymer is prepared by isolating a molecular weight fraction from a starting polymeric material which has repeat units of Formula 1. The polymeric molecular weight fractions disclosed therein may also be useful as materials in the present invention. Accordingly, the contents of that patent application are incorporated herein by reference.

Examples of further preferred polytriarylamine, which may be substituted by at least one optionally substituted long chain, linear, branched or cyclic carbyl-derived group,

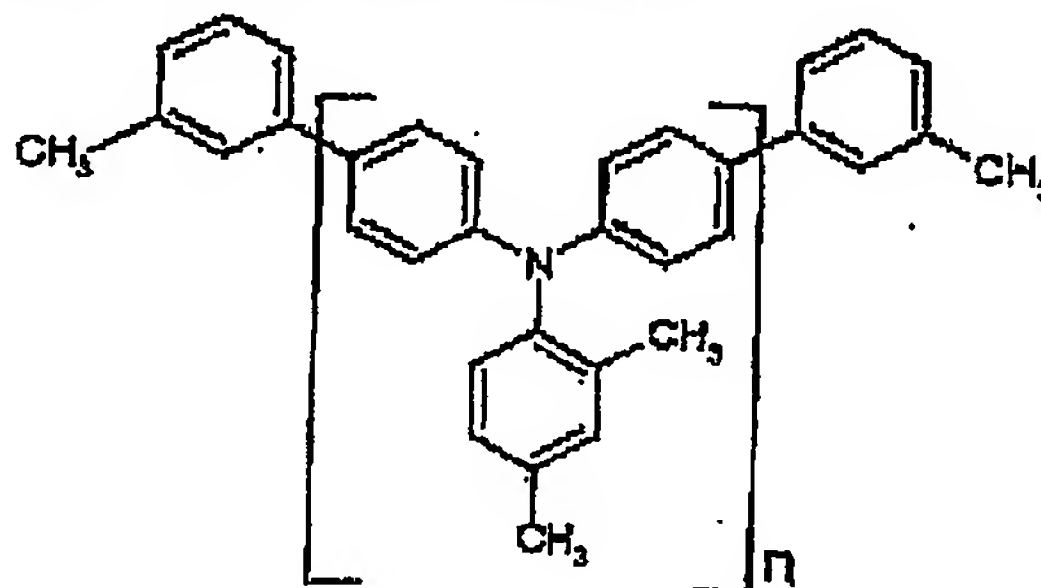
SMC 60569

8

i.e. C_8 or longer, preferably an optionally substituted long chain hydrocarbyl-derived group, most preferably alkyl or alkoxy, are given by formulae 1A and 1B:



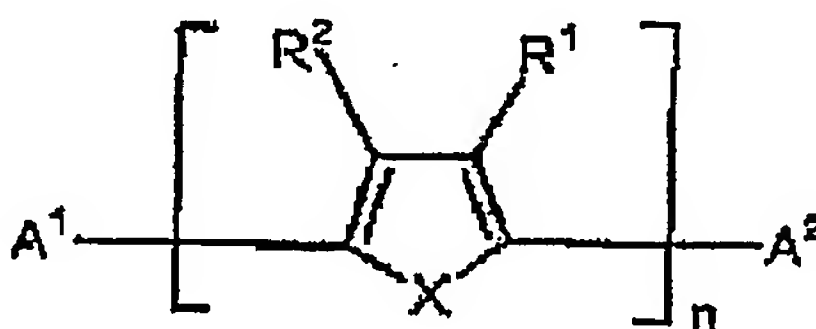
Formula 1A



Formula 1B

Examples of other suitable polymeric materials than polyarylamines are now described.

Conjugated oligomeric and polymeric heterocyclic semiconductors may comprise a repeat unit of an optionally substituted 5 membered ring and terminal groups A^1 and A^2 as shown in Formula 2:



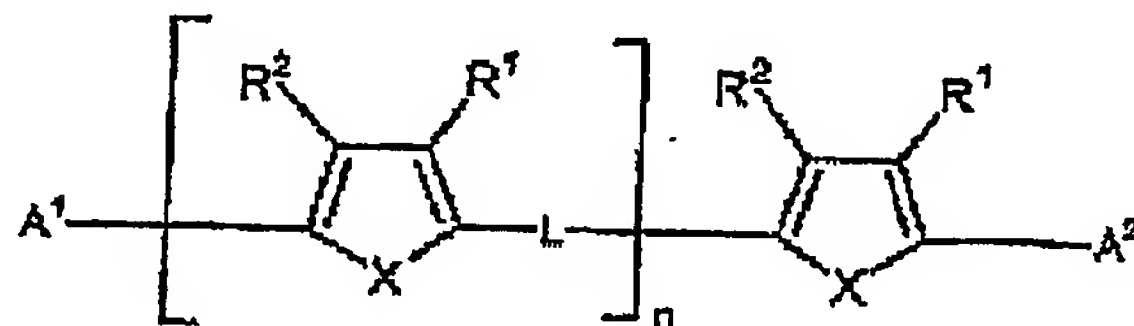
Formula 2

in which X may be Se, Te or preferably O, S, or $-N(R)-$ where R represents H, optionally substituted alkyl or optionally substituted aryl; R^1 , R^2 , A^1 and A^2 may be independently H, alkyl, alkoxy, thioalkyl, acyl, aryl or substituted aryl, a fluorine atom, a cyano group, a nitro group or an optionally substituted secondary or tertiary alkylamine or arylamine $-N(R^3)(R^4)$, where R^3 and R^4 are each independently H, optionally substituted alkyl, optionally substituted aryl, alkoxy or polyalkoxy groups. The alkyl and aryl groups may be optionally fluorinated. The alkyl and aryl groups represented by R^1 , R^2 , R^3 , R^4 , A^1 and A^2 may be optionally fluorinated. The number of recurring units in the conjugated oligomer of Formula 2 is represented by an integer n , where n is preferably 2 to 14. Preferred oligomers have $X = S$, R^1 and $R^2 = H$ and A^1 and $A^2 =$ optionally substituted C_{1-12} alkyl groups, examples of especially preferred compounds being A^1 and $A^2 = n$ -hexyl and where $n=4$, alpha-omega-n-hexylquaterthienylene (alpha-omega 4T), $n=5$, alpha-omega-n-hexylpentathienylene (alpha-omega -5T), $n=6$, alpha-omega-n-hexylhexathienylene (alpha-omega-6T), $n=7$, alpha-omega-n-hexylheptathienylene (alpha-omega-7T), $n=8$, alpha-omega-n-hexyloctathienylene (alpha-omega-8T), and $n=9$, alpha-omega-n-hexylnonathienylene (alpha-omega-9T).

SMC 60669

9

Oligomers containing a conjugated linking group may be represented by Formula 3:



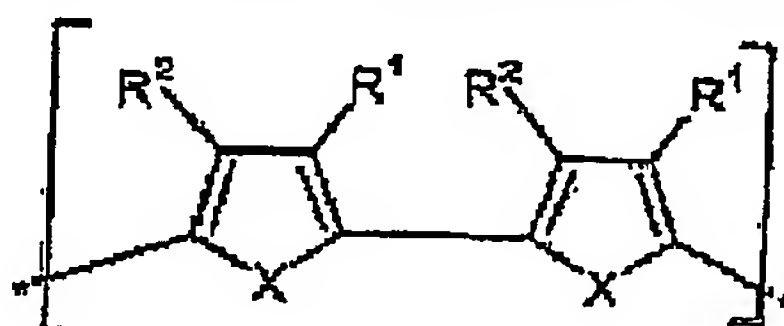
Formula 3

5

in which X may be Se, Te, or preferably O, S, or -N(R)-, R is as defined above; R^1 , R^2 , A^1 and A^2 as defined above for Formula 2. Linking group L represents -C(T_1)=C(T_2)-, -C≡C-, -N(R')-, -N=N-, (R')=N-, -N=C(R')- with T_1 and T_2 each independently representing H, Cl, F, -C≡N or lower alkyl groups particularly C_{1-4} alkyl groups; R' represents H, optionally substituted alkyl or optionally substituted aryl. The alkyl and aryl groups may be optionally fluorinated,

10

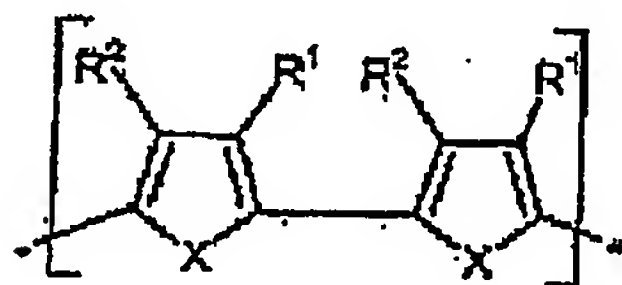
Polymers may have repeat units of the general Formula 4:



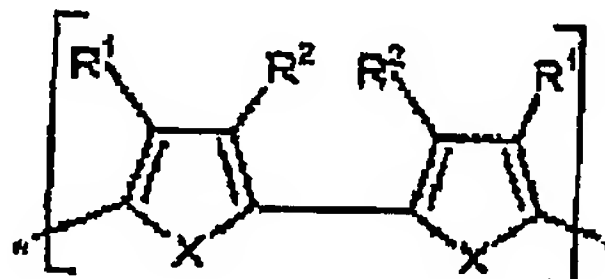
Formula 4

15

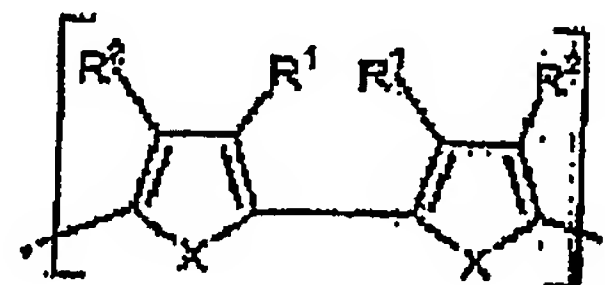
in which X, R^1 and R^2 are defined as above. The sub units may be polymerised in such a way as to give a regio regular or a regio random polymer comprising repeat units as shown in Formulae 4 to 6:



Formula 4



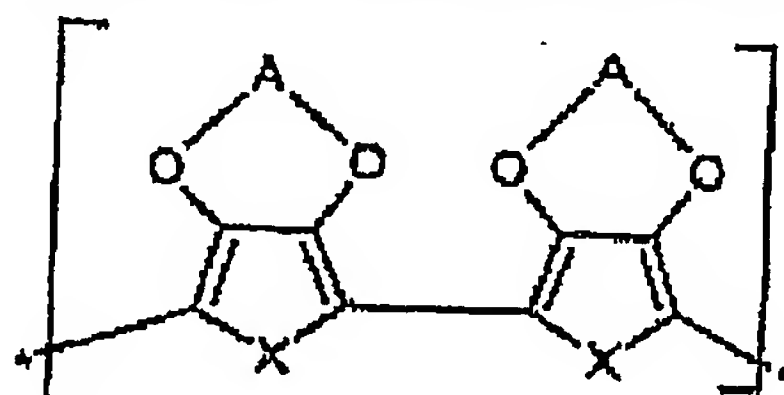
Formula 5



Formula 6

20

Polymers may have repeat units of the general Formula 7:



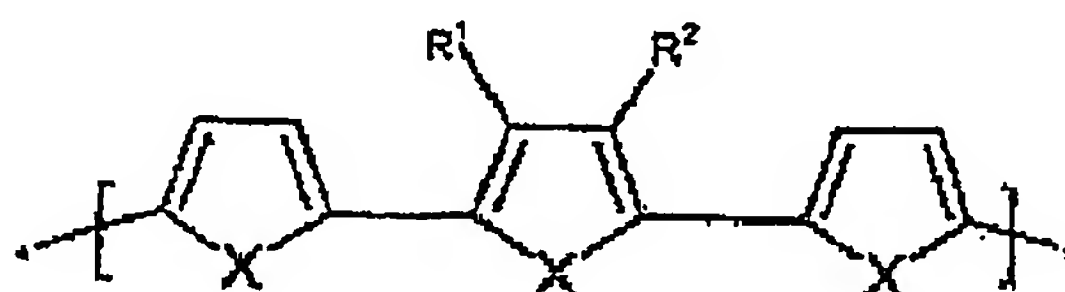
Formula 7

SMC 60569

10

in which X is as defined above and the bridging group A is optionally fluorinated C_{1-8} alkyl, for example poly(3,4-ethylenedioxy)thiophene-2,5-diyl and poly(3,4-trimethyldioxy)thiophene-2,5-diyl.

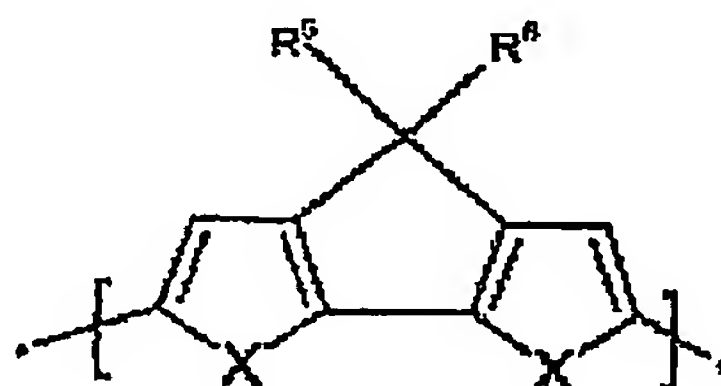
Polymers may have repeat units of general Formula 8:



Formula 8

in which X, R^1 and R^2 are defined as above. Specific examples are where one of R^1 or R^2 is an alkoxide of general formula $C_nH_{2n+1}O-$, and the other of R^1 or R^2 is H, poly(β' -dodecyloxy- α,α' - α,α'' terthienyl) i.e. polyDOT₃.

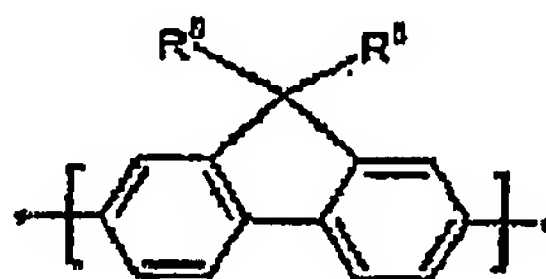
Polymers may have repeat units of general Formula 9:



Formula 9

in which X is as defined above; R^5 and R^6 may be independently H, alkyl, aryl or substituted aryl. The alkyl and aryl groups may be optionally fluorinated.

Polymers may have repeat units of general Formula 10 in which R^5 and R^6 are as defined in Formula 9:



Formula 10

Copolymers comprising repeat units as above described and also other repeat units comprising two or more of the repeat units could be used.

The non-polymeric material preferably is a small molecule. The non-polymeric material preferably comprises a conjugated aromatic molecule containing three to twelve aromatic rings. Preferred molecules contain 5, 6 or 7 membered aromatic rings, especially 5 or 6 membered aromatic rings.

SMC 60569

11

Each of the aromatic rings of the preferred non-polymeric material may optionally contain one or more hetero atoms selected from Se, Te, P, Si, B, As, N, O or S; preferably from N, O or S. The rings may be optionally substituted, e.g. with optionally substituted alkyl, alkoxy, polyalkoxy, thioalkyl, acyl, aryl or substituted aryl groups, a fluorine atom, a cyano group, a nitro group or an optionally substituted secondary or tertiary alkylamine or arylamine $-N(R^3)(R^4)$, where R^3 and R^4 each independently is H, optionally substituted alkyl, optionally substituted aryl, alkoxy or polyalkoxy groups. The alkyl and aryl groups may be optionally fluorinated.

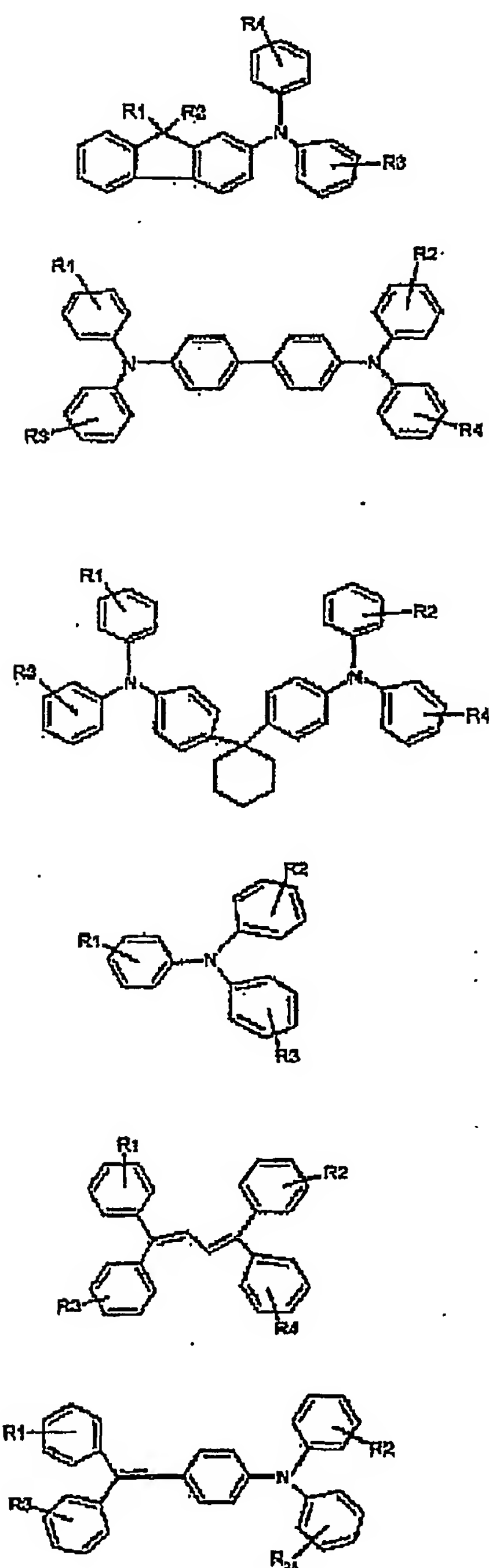
The aromatic rings of the preferred non-polymeric material may be optionally fused or may be linked with a conjugated linking group such as $-C(T_1)=C(T_2)-$, $-C\equiv C-$, $-N(R')-$, $-N=N-$, $(R')=N-$, $-N=C(R')-$. T_1 and T_2 each independently represent H, Cl, F, $-C\equiv N$ or lower alkyl groups particularly C_{1-4} alkyl groups; R' represents H, optionally substituted alkyl or optionally substituted aryl. The alkyl and aryl groups may be optionally fluorinated.

Preferred non-polymeric materials may be small molecules selected from triarylamine, fluorene, butadiene derivatives, carbazole, stilbene and spiro, enamine, oxadiazole, oxazole, pyrazoline and triarylmethane compounds and derivatives. Specific examples for small molecules include 2-(4-biphenyl)-5-(tert-butylphenyl)-1,2,4-oxadiazole (PBD), PTCDA, 4,4'-di(N-carbazolo)diphenyl (CBP). Examples of classes of preferred non-polymeric material are given below where the generic structure of a class is given on the left and next to it on the right is given a preferred example from that class. The substituents R_1-R_4 are independently selected carbyl-derived groups, preferably hydrocarbyl-derived groups, e.g. alkyl, alkoxy, polyalkoxy, thioalkyl, acyl, and aryl groups, all of which are optionally substituted, or other substituents e.g. halo, hydroxy, amino, cyano or nitro. Preferably, R_1-R_4 are independently selected from alkyl, alkoxy or aryl, each of which may be optionally substituted. The ability to change the substitution in the molecules makes it easy to tune the HOMO level so that it aligns well with, e.g., the HOMO level of the EL layer.

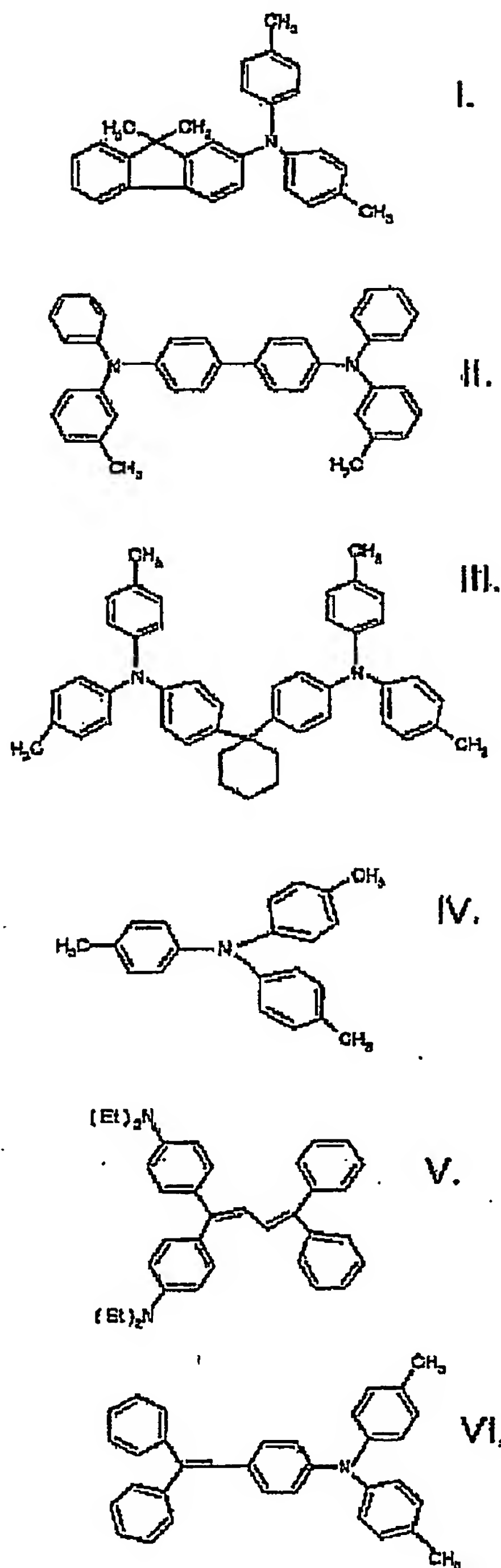
SMC 60569

12

General structure



Example



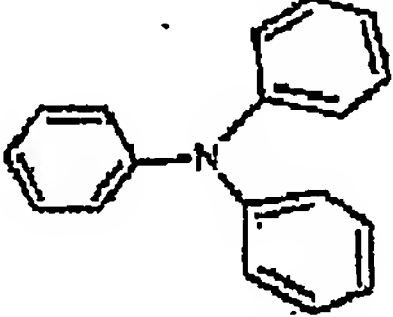
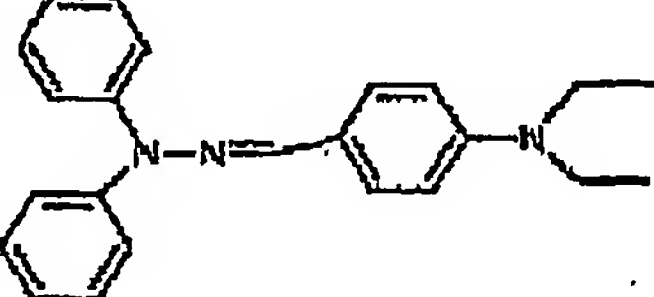
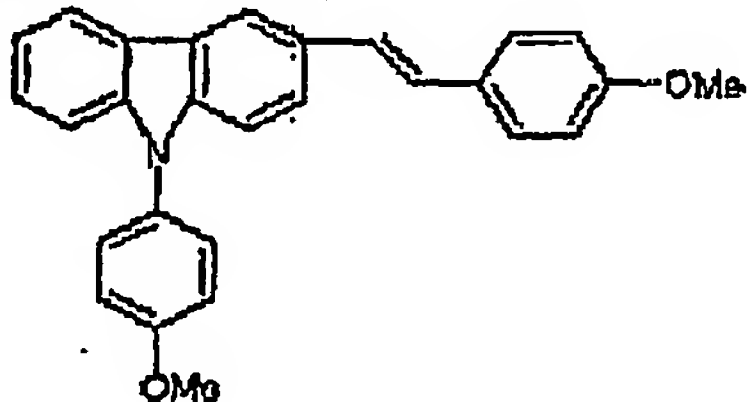
5 A preferred composition comprises a polyarylamine, more preferably a polytriarylamine, and a non-polymeric conjugated aromatic molecule containing three to twelve aromatic rings. A preferred polytriarylamine is one which contains repeat units of

SMC 60569

13

- Formula 1, 1A or 1B above. Preferably, the polytriarylamine is substituted by at least one optionally substituted long chain carbyl-derived group, i.e. C_8 or longer, preferably an optionally substituted long chain hydrocarbyl-derived group, most preferably alkyl or alkoxy. The value of n may be from 3 to 20,000 but most preferably is from 6 to 100. The
- 5 polymeric material of the invention is preferably polydisperse. Preferably the polydispersity is from 1.1 to 5. More preferably, the polydispersity is from 1.1 to 3. Preferred non-polymeric conjugated aromatic molecules for use with the polytriarylamine are those of Formulas I-VI above. Other preferred examples of polymeric and non-polymeric materials are given in Table 1 below, together with data for the HOMO levels (i.p.) of the materials,
- 10 Derivatives, including substituted forms of the materials in Table 1 may be used. Preferably, the particular preferred compositions described above form a hole injection layer and/or a hole transport layer.

Table 1

Hole transport small molecules:	HOMO (i.p.)	Reference for examples
V (see above) (BD)	5.12 eV	T. Kitamura and Y. Chigono, 1991 Proc. of SPSE, p276
IV (see above) (TPTA)	5.5 eV	T. Kitamura and M. Yokohama, 1990, J. of Imaging Sci. 34 (5) p197
II (see above) (TPD)	5.35 eV	
 (TPA)	5.7 eV	T. Kitamura and M. Yokohama, 1990, J. of Imaging Sci. 34 (5) p197
 DEH	5.36 eV	T. Kitamura and Y. Chigono, 1991 Proc. of SPSE, p276
	5.72 eV	T. Kitamura and Y. Chigono, 1991 Proc. of SPSE, p276

SMC 60569

14

 α -NPB		
 β -NPB		
Hole transport polymers:		
PTAA (polytriarylamine)		WO 99/32537 and WO 00/78843
Polyfluorene variants		Examples in US 6,309,763 (Dow)
polyspiros		Examples in US 6,476,265 (Covion)
Carbazoles, e.g. Polyvinylcarbazole (PVK)		

It can be seen that the HOMOs of the examples of both the polymeric and non-polymeric materials in Table 1 cover a significant range between about 4.8 and about 5.7 eV and thus either type of material can be used to match to the w.f. of the anode or HOMO of the EL layer or HOMO of another layer. Typically, the polymeric material is used to match to the anode and the non-polymeric material is used to match to the EL layer or other intermediate layer.

Advantageously, the composition of the present invention exhibits the following properties: high carrier mobility, compatibility with binders, improved solubility, high durability and/or high resistivity undoped. The composition is highly effective for use in EL devices. It has superior film forming properties, particularly when n is greater than 7 and the polymeric material and/or non-polymeric material are substituted with one or more optionally substituted long chain carbyl-derived groups (preferably alkyl or alkoxy), i.e. C_6 or longer.

Advantageously, the composition may be easily and cheaply deposited on the device since the composition is solution coatable, i.e. it may be readily deposited from solution. Preferably, the composition is applied by a solution coating technique. Preferably, the composition is laid down in a film form. The composition may be laid down in a film form, which can be optionally patterned or structured, by a variety of coating or printing techniques including, but not limited to, dip coating, roller coating, reverse roll

SMC 60569

15

coating, bar coating, spin coating, gravure coating, lithographic coating (including photolithographic processes), ink jet coating (including continuous and drop-on-demand, and fired by piezo or thermal processes), screen coating, spray coating and web coating. Other layers in the device may be deposited by solution coating or by conventional vapour deposition.

In fabrication of a device according to the invention, other than a so-called "top-emission device" described separately below, when used as a HIL or HTL the layer comprising the composition of the invention may be solution coated onto the anode or onto a separate HIL provided by known means on the anode, followed by deposition of subsequent layers, including the EL layer, by solution coating or by conventional vapour deposition.

According to another aspect of the invention, there is provided a method of forming the electroluminescent device, which method comprises depositing from a solution the layer comprising the composition. Optionally, the method further comprises depositing at least one other layer, e.g. the EL layer, by vapour deposition or deposition from solution.

As mentioned above, the composition of the present invention has both excellent film forming ability and high mobility. The layer thickness may be from 10 nm to 500 nm, preferably from 20 to 500 nm. Preferably, the layer comprising the composition is applied at high thickness, preferably greater than 40 nm, more preferably greater than 60 nm, still more preferably greater than 100 nm, and most preferably greater than 200 nm. Preferably the thickness is up to 500 nm, more preferably up to 400 nm, still more preferably up to 300 nm, whilst still achieving high yield. Such thick layers have been found to provide numerous advantages, for example, enhanced device lifetime, reproducibility, yield and luminescence. Without being bound by any theory, it is believed that such thick layers improve the device yield by making the structure less sensitive to substrate defects. For example, when the layer is used as a HIL, the thick layer yields particular improvement when the layer is coated directly onto an indium tin oxide (ITO) anode as it is believed that it helps to eliminate the roughness of ITO better than conventional injection layers such as PANi or PEDOT. It has been found that thick layers formed by the composition of the invention improve device lifetime. Again, without being bound by any theory, it is believed that this is also due to reducing the effects of surface defects. In particular, the thick layer may reduce shorting effects and local spots at the electrode, thereby increasing the lifetime of the EL layer. The composition has been found to be particularly useful in this regard for blue emitting EL materials.

The composition preferably has a carrier (hole or electron) mobility greater than $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is an enabling factor in the fabrication of such thick layers. The high mobility of the composition used in the present invention also enables the drive voltage to be kept relatively low for high luminescent efficiencies. The high mobility of the composition of the invention means that the potential drop across the layer comprising the

SMC 60569

16

composition can be very small. Conventional PANi or PEDOT Injection layers have low conductivity to avoid "crosstalk" between neighbouring pixels in display devices. However, a thick layer of the composition of the invention can effectively perform this function due to its unipolar nature.

5 The composition of the invention advantageously also enables a high quality ohmic interface between the composition and the electrode, e.g. anode, such as an ITO anode. This in turn yields improvements in device lifetime since non-ohmic contact is thought in part to be responsible for hot spots due to localised build up of electric field at the electrode resulting in break down of the adjacent injection layer. By reducing the
10 potential drop in the immediate vicinity of the anode or cathode, the build-up of local fields is reduced and the contact becomes more reliable which in turn leads to improved device lifetime. It has not previously been easy to find appropriate charge transport materials that allow for an ohmic contact. It has previously been tried to employ doped transport layers. However, this such doping is difficult to implement in a real manufacturing process
15 (a sequence of many layers is required) and attractive lifetime has not yet been demonstrated.

ITO is a highly preferred anode material due to its transparency, high conductivity and availability on glass or polymer substrates. ITO has a workfunction between 4.8-5.0 eV. In a preferred embodiment, the composition is suitable for use as a hole injection or
20 transport layer and has an HOMO energy level (ionisation potential) close to this value, preferably in the range 4.8-5.2 eV. One of the HOMO levels in the composition should have an energy within ± 0.2 eV of the w.f. of the anode. The polymeric material of the composition is suited for this purpose as there is a wide range of such materials with different energy HOMO levels. One preferred polymeric material is a polyarylamine, e.g.
25 described by general formula 1. Thus, holes can be injected into the organic layer comprising the composition unhindered. The HOMO level of EL materials is dependent on their colour. The HOMO is typically higher than the workfunction of ITO for example, often as high as 5.5 eV. This is especially the case for blue EL materials. The present invention enables a single HIL comprising the composition to match the EL material as well as the
30 anode. One of the HOMO levels in the composition should have an energy within ± 0.2 eV of the HOMO of the EL layer. The non-polymeric material of the composition is ideally suited for this purpose as there is a wide range of such materials with different HOMO levels. For example, TPD has a HOMO of 5.35 eV which may be used with an EL layer having a HOMO level of 5.5 eV for example. The structure of TPD is shown by formula II
35 above.

Alternatively, the HOMO level of a polymeric material may be used for matching to the HOMO of the EL layer. For example polyvinylcarbazole (PVK) has a HOMO of approximately 5.6 eV, well suited to the HOMO position of many blue EL layers. A non-polymeric material of lower ionisation potential may be then used to match the
40 workfunction of the anode.

SMC 60569

17

Conventionally, as shown in Figure 1 (prior art), materials such as PANI or PEDOT are used for the role of the HIL. A separate HTL is typically employed between the PANI or PEDOT layer and the EL layer. We have found that the composition of the invention can effectively perform both the injection function of the PANI or PEDOT and the role of the separate HTL due to having one or more HOMO energy level(s)/ionisation potential aligned with the anode work function as well as having one or more HOMO energy level(s)/ionisation potential suitably aligned with the HOMO level of the EL layer. The composition, applicable in thick layers e.g. from a solution, also has excellent surface levelling ability. Thus, in one advantageous embodiment, the invention provides a HIL comprising the composition, i.e. without need for, e.g., PANI or PEDOT. In a further advantageous embodiment, the invention provides an EL device in which there is only one organic layer between the anode and the (organic) EL layer, as shown in Figure 4. This single organic layer (HIL) comprising the composition between the anode and the EL layer greatly simplifies the device and processing thereof. In addition, there is no loss of efficiency; indeed, efficiency in many cases is improved. This solves the problem in the prior art of needing multiple organic layers (HIL, HTLs etc.). In Figure 4 the layers on the electron injection and transport side have been shown in dotted outline to indicate that the number of layers on this side may be varied. In an advantageous embodiment a composition according to the invention is also used as a single layer between the EL layer and the cathode.

It will be appreciated that other embodiments may exist wherein, if desired, the composition of the invention may be used with a separate HIL, e.g. comprising PANI or PEDOT, whilst still providing benefits. Thus, in another embodiment, the invention provides a HTL comprising the composition.

In another embodiment, there may be a HIL comprising the composition of the invention and, in addition, one or more HTL(s) comprising the composition of the invention, the composition in each of the HIL and HTL(s) being independently optimised in terms of its ionisation potentials, e.g., for matching to the anode and EL layer respectively.

The composition of the invention, when used in place of certain conventional EL device materials, e.g. PANI or PEDOT, can yield improvements in transparency and colour rendition, especially with blue emitting EL devices. This is due to the composition being substantially transparent or "white". The composition can be substantially amorphous.

It has been found that hole carrying compositions of the invention are better at blocking electrons than conventional materials, e.g. PANI or PEDOT, thus leading to improvements in device efficiency.

The composition of the invention has been found to be more chemically stable than conventional HIL material such as PANI or PEDOT. Conventional PANI and PEDOT materials for example are acid doped and possess counter-ions which, with time, migrate into adjacent layers and cause a deterioration in device performance. The composition of

SMC 60569

18

the invention, however, does not need counter-ions, thus eliminating the problem. This is especially advantageous for triplet^{emitting} devices.

The compositions of the invention have a relatively high glass transition temperature (T_g), which leads to improved stability.

5 The composition may be used in conjunction with a binder resin to further improve film formation and/or adjust viscosity for improving solution coatability. The binder may also be optionally crosslinked for improved stack integrity of layers as described in more detail below. A binder is preferred for an EL device wherein all of the organic layers are solution coated.

10 Preferred binders are electrical insulators. Preferred binders include, without limitation, at least one of polyamide, polyurethane, polyether, polyester, epoxy resin, polyketone, polycarbonate, polysulphone, vinyl polymer (for example polyvinylketone and/or polyvinylbutyral), polystyrene, polyacrylamide, copolymers thereof (such as aromatic copolymeric polycarbonate polyesters) and/or compositions thereof.

15 Those binders disclosed in the patent application WO 99/32537 of the applicant, particularly at pages 24 and 25 of that application as published, are useful for the present invention and that disclosure is incorporated herein by reference. In addition, those binders claimed in and described in the patent application WO 02/45184 of the applicant, particularly at pages 3, 4, 8-11 of that application as published, including those listed in
20 Tables 1 and 2 therein, are useful for the present invention and that disclosure is also incorporated herein by reference.

To improve stack integrity between the layer comprising the composition and adjacent layers, the layer comprising the composition optionally may be crosslinked. The crosslinking may be achieved by crosslinking of the composition, e.g. by means of a
25 crosslinkable functionality in the polymer, and/or by crosslinking of the binder resin where present, for example as disclosed in WO 02/45184.

Advantageously, the composition of the invention provides routes to novel device structures by enabling depositing of the transparent electrode on top of an OLED stack for a 'top emission device' preferred in some active matrix display configurations for
30 increased luminance and resolution. It is very desirable for the deposition of the transparent top electrode to use fast processes such as sputtering. For example for depositing ITO. However, these processes are likely to damage the vulnerable active organic layers due to the high kinetic energy of the particles deposited. A thick hole transport layer for example, formed by the composition of the invention, provides
35 protection and thus allows for a robust and commercially viable manufacturing process (high yield) without affecting the device performance (efficiency, driving voltage, lifetime).

As mentioned, the invention may equally be used to provide improved electron injection/transport layers (EIL/ETL). The EIL/ETL may contain non-polymeric or polymeric material comprising Alq3, oxidiazol, triazin, bathophenanthroline units. Further examples
40 of suitable transport materials for use in the composition according to the invention are

SMC 60569

19

given in Table 2 below. Also, suitable electron transport materials may comprise be polymers comprising triazine, oxidiazole, phenylene, vinylene, cyano-phenylene and cyano-vinylene units. In one embodiment, the LUMO level of one of the materials is such that it matches close to (preferably within ± 0.2 eV of) the cathode workfunction and the

5 LUMO level of one of the other materials is such that it matches close to (preferably within ± 0.2 eV of) the LUMO level of the EL layer.

SMC 60569

20

Table 2

Electron transport small molecules:
AIQ3
Bathophenanthroline
PBD
CuPc
PTCDA
Electron transport polymers:
Polymeric triazines
Cyano-PPVs

Examples of EL materials with which the composition of the invention may be used are given in Table 3 below.

5

Table 3

Luminescent materials:
MEH-PPV
Alkoxy PPV
Polyfluorenes

PPV=polyphenylenevinylene

10 Various terms used herein are now defined. The term 'carbyl-derived' as used herein denotes any monovalent or multivalent organic radical moiety which comprises at least one carbon atom either without any non-carbon atoms (e.g. $-C\equiv C-$), or optionally combined with at least one other non-carbon atom (e.g. alkyl, carbonyl etc.). The non-carbon atom(s) may comprise any elements other than carbon (including any chemically possible compositions or combinations thereof) that together with carbon can comprise an organic radical moiety. Preferably the non-carbon atom is selected from at least one hydrogen and/or heteroatom, more preferably from at least one: hydrogen, phosphorus, halo, nitrogen, oxygen and/or sulphur, most preferably from at least one hydrogen, nitrogen, oxygen and/or sulphur. Carbyl-derived groups include all chemically possible combinations in the same group of a plurality (preferably two) of the aforementioned

15

20

SMC 60569

21

carbon and/or non-carbon atom containing moieties (e.g. alkoxy and carbonyl) if directly attached to each other represent an alkoxy carbonyl radical).

Throughout the description and claims of this specification, the words "comprise" and "contain" and variations of the words, for example "comprising" and "comprises", mean "including but not limited to", and are not intended to (and do not) exclude other components.

It will be appreciated that variations to the foregoing embodiments of the invention can be made while still falling within the scope of the invention. Each feature disclosed in this specification, unless stated otherwise, may be replaced by alternative features serving the same, equivalent or similar purpose. Thus, unless stated otherwise, each feature disclosed is one example only of a generic series of equivalent or similar features.

All of the features disclosed in this specification may be combined in any combination, except combinations where at least some of such features and/or steps are mutually exclusive. In particular, the preferred features of the invention are applicable to all aspects of the invention and may be used in any combination. Likewise, features described in non-essential combinations may be used separately (not in combination).

It will be appreciated that many of the features described above, particularly of the preferred embodiments, are inventive in their own right and not just as part of an embodiment of the present invention. Independent protection may be sought for these features in addition to or alternative to any invention presently claimed.

Example

An example of a composition comprises a polytriarylamine (PTAA) described by general formula 1. A PTAA polymer of formula 1A is used and a HOMO level of 5.1 eV is obtained. The PTAA polymer is mixed with TPD (a small molecule hole transporting material which has a HOMO of 5.35 eV) at 50/50% by weight in a solvent which also contains a binder resin. The resulting solution is solution coated onto an ITO anode of work function 5.0 eV. An EL layer having a HOMO of 5.5 eV and subsequent layer(s) including the cathode are then deposited on top. The solution coated layer remains amorphous and has two HOMO layers, matching (i.e. within +/- 0.2 eV) the ITO workfunction and the HOMO of the EL layer respectively. Thus the EL device formed has only one HIL layer as shown in Figure 4. In contrast, currently this energy level matching is done by multilayer deposition of different HTL in addition to PANi or PEDOT injection layers as shown for example in Figure 1. Additionally, the composition, by correct matching and ohmic contact with ITO, results in a long lifetime as it reduces the possibility of large fields across the interface with the anode. ITO is a high conductivity layer and is therefore capable of producing local breakdown when such fields are present.

SMC 60569

22

Claims

5 1. An electroluminescent device having an anode, a cathode and one or more organic layers between said anode and said cathode, at least one of said organic layers comprising an organic electroluminescent material, wherein at least one of said organic layers comprises a composition having at least two HOMO energy levels and/or at least two LUMO energy levels.

10 2. An electroluminescent device as claimed in claim 1 wherein the energy of the at least two HOMO energy levels and/or at least two LUMO energy levels are each within ± 0.2 eV of an electronic energy level in an adjacent layer.

15 3. An electroluminescent device as claimed in claim 2 wherein the energy of at least one of the HOMO levels is within ± 0.2 eV of the work function of the anode or the energy of at least one of the LUMO levels is within ± 0.2 eV of the work function of the cathode.

20 4. An electroluminescent device as claimed in either claim 2 or 3 wherein the energy of at least one of the HOMO levels is within ± 0.2 eV of the HOMO level of the EL layer or the energy of at least one of the LUMO levels is within ± 0.2 eV of the LUMO level of the EL layer.

25 5. An electroluminescent device as claimed in any one preceding claim wherein the energy difference between the at least two HOMO energy levels and/or between the at least two LUMO energy levels is greater than 0.1 eV.

30 6. An electroluminescent device as claimed in claim 5 wherein the energy difference is greater than 0.2 eV.

35 7. An electroluminescent device as claimed in any one preceding claim wherein the composition comprises at least two charge transport materials, the energies of the HOMOs of the materials and/or the energies of the LUMOs of the materials being different.

40 8. An electroluminescent device as claimed in claim 7 wherein the at least two charge transport materials comprise at least one polymeric material and at least one non-polymeric material.

9. An electroluminescent device as claimed in claim 7 or 8 wherein the at least two charge transport materials are organic semiconductors.

SMC 60569

23

10. An electroluminescent device as claimed in any one preceding claim wherein the materials are positive charge (hole) carrier materials.
- 5 11. An electroluminescent device as claimed in claim 10 wherein the layer comprising the composition is a hole injection layer (HIL) and/or a hole transport layer (HTL).
- 10 12. An electroluminescent device as claimed in any one of claims 1 to 9 wherein the materials are negative charge (electron) carrier materials.
- 15 13. An electroluminescent device as claimed in claim 12 wherein the layer comprising the composition is an electron injection layer (EIL) and/or electron transport layer (ETL).
- 20 14. An electroluminescent device as claimed in any one preceding claim wherein the device comprises only one organic layer such that the organic layer comprising the composition of the invention is the same layer as the layer comprising the electroluminescent material.
- 25 15. An electroluminescent device as claimed in any one of claims 1 to 13 wherein the device comprises two or more organic layers, the organic layer comprising the composition of the invention being different to the layer comprising the electroluminescent material.
- 30 16. An electroluminescent device as claimed in any one preceding claim wherein the polymeric material has a HOMO level matched close in energy to, preferably within ± 0.2 eV of, the w.f. of the anode and/or the non-polymeric material has a HOMO level matched close in energy to, preferably within ± 0.2 eV of, the HOMO level of the EL layer.
- 35 17. An electroluminescent device as claimed in any one of claims 1 to 15 wherein the non-polymeric material has a HOMO level matched close to, preferably within ± 0.2 eV of, the w.f. of the anode and/or the polymeric material has a HOMO level matched close to, preferably within ± 0.2 eV of, the HOMO level of the EL layer.
- 40 18. An electroluminescent device as claimed in any one preceding claim wherein the energies of at least one of the HOMO levels of the composition lies in the range from about 4.9 to about 5.7 eV.

SMC 60569

24

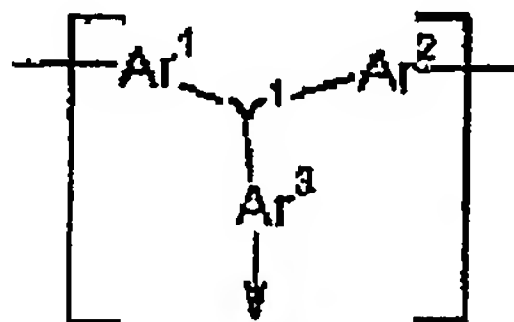
- 5
19. An electroluminescent device as claimed in any one preceding claim wherein the at least one polymeric material and the at least one non-polymeric material are mixed in a ratio from 10:90 to 90:10 % by weight.
20. An electroluminescent device as claimed in any one preceding claim wherein the number of repeat units which may be present per molecule in the polymeric material ('n') is from 3 to 20,000.
- 10
21. An electroluminescent device as claimed in any one preceding claim wherein the polymeric material is polydisperse.
22. An electroluminescent device as claimed in any one preceding claim wherein the polymeric material and/or the non-polymeric material is substituted by at least one optionally substituted long chain, linear, branched or cyclic carbyl-
15 derived group which is C_n or longer.
23. An electroluminescent device as claimed in claim 22 wherein the group comprises optionally substituted alkyl or alkoxy.
- 20
24. An electroluminescent device as claimed in any one preceding claim wherein the at least one polymeric material is selected from compounds and derivatives of compounds of the following list : conjugated organic (including hydrocarbon) polymers selected from polyarylamine, polytriarylamine, polyacene, polyphenylene, poly(phenylene vinylene), polyfluorene including oligomers of those conjugated hydrocarbon polymers; oligomeric para substituted phenylenes including p-quaterphenyl (p-4P), p-quinquephenyl (p-5P), p-sexiphenyl (p-6P); conjugated heterocyclic polymers including polythiophene, poly(3-substituted thiophene), poly(3,4-bisubstituted thiophene), polybenzothiophene, polyisothianaphthene, poly(N-substituted pyrrole), poly(3-substituted pyrrole), poly(3,4-bisubstituted pyrrole), polyfuran, polypyrrolidine, poly-1,3,4-oxadiazoles, polyisothianaphthene, poly(N-substituted aniline), poly(2-substituted aniline), poly(3-substituted aniline), poly(2,3-bisubstituted aniline), polyazulene, polypyrene; pyrazoline compounds; polyselenophene; polybenzofuran; polyindole; polypyridazine; benzidine compounds; stilbene compounds; triazines; carbazole polymers ; butadiene polymers.
- 25
- 30
- 35
- 40
25. An electroluminescent device as claimed in claim 24 wherein the polymeric material is a polyarylamine, preferably a polytriarylamine.

SMC 60569

25

26. An electroluminescent device as claimed in claim 25 wherein the polytriarylamine has repeat units of formula 1:

5 Formula 1:



wherein:

10 Y^1 represents, independently if in different repeat units, N, P, S, As and/or Se, preferably N;

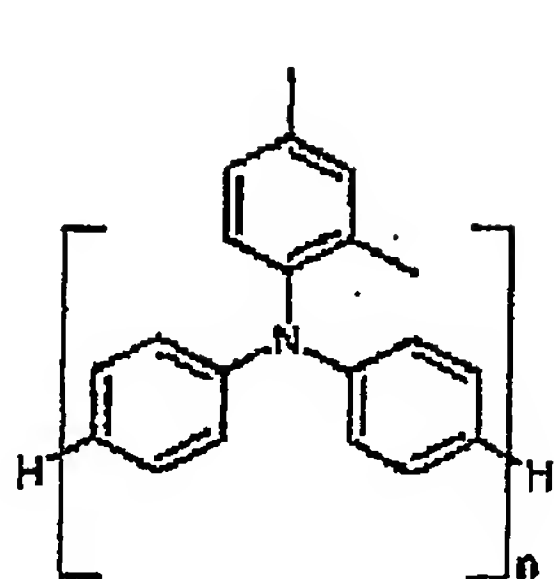
15 Ar^1 and Ar^2 are aromatic groups and Ar^3 is present only if Y^1 is N, P, or As in which case it too is an aromatic group; wherein Ar^1 and Ar^2 are the same or different and represent, independently if in different repeat units, a multivalent (preferably bivalent) aromatic group (preferably mononuclear but optionally polynuclear) optionally substituted by at least one optionally substituted C_{1-10} carbyl-derived groups and/or at least one other optional substituent; and Ar^3 represents, independently if in different repeat units, a mono or multivalent (preferably bivalent) aromatic group (preferably mononuclear but optionally polynuclear) optionally substituted by at least one optionally substituted C_{1-10} carbyl-derived group and/or at least one other optional substituent.

20

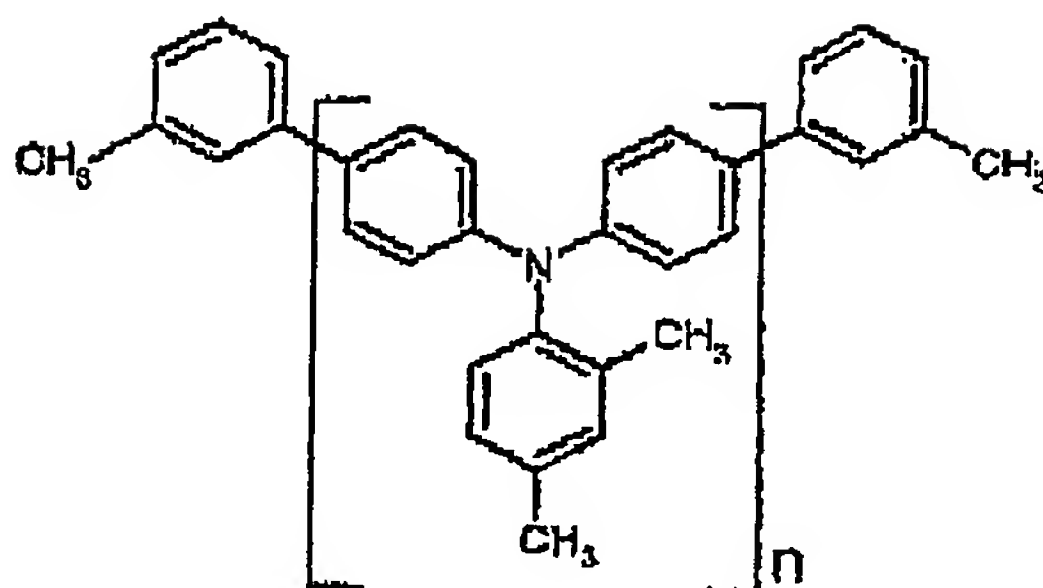
27. An electroluminescent device as claimed in claim 26 wherein the polytriarylamine has a formula 1A or 1B:

SMC 60569

26



Formula 1A

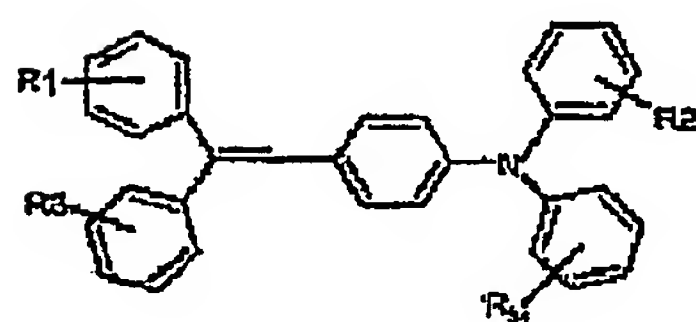
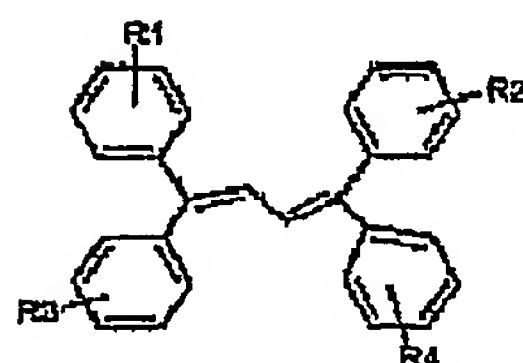
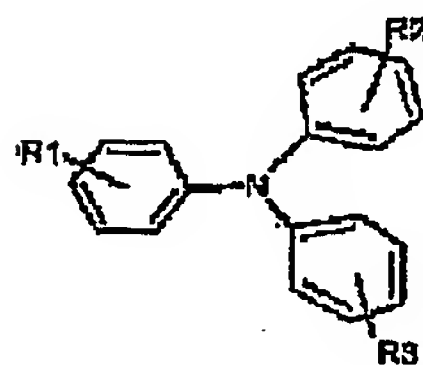
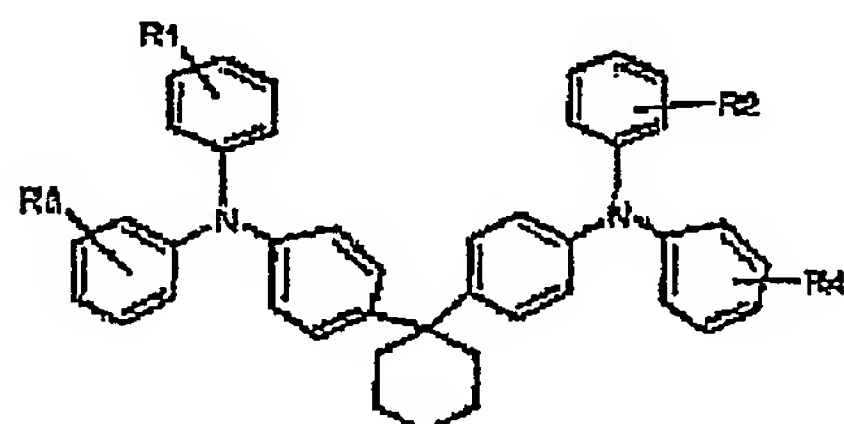
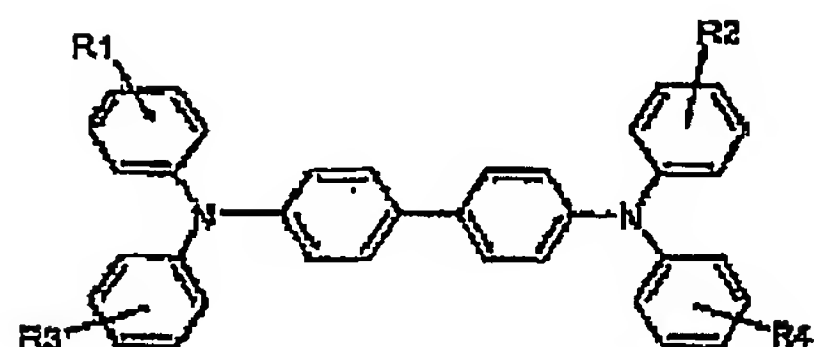
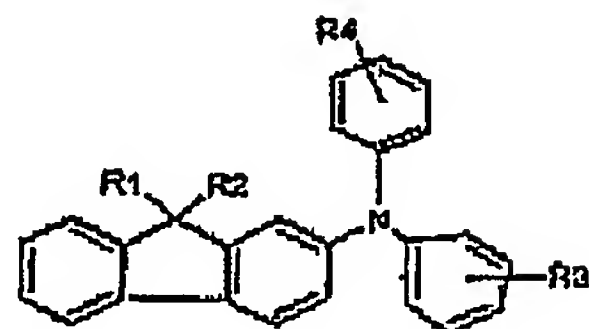


Formula 1B

- 5
28. An electroluminescent device as claimed in any one of claims 1 to 23 wherein the polymeric material is an electron transporting material selected from polymers comprising triazine, oxidiazole, phenylene, vinylene, cyano-phenylene and cyano-vinylene units.
- 10
29. - An electroluminescent device as claimed in any one preceding claim wherein the non-polymeric material comprises a conjugated aromatic molecule containing three to twelve aromatic rings.
- 15
30. An electroluminescent device as claimed in any one preceding claim wherein the non-polymeric material is selected from triarylamine, fluorene, butadiene, carbazole, stilbene and spiro compounds and derivatives.
- 20
31. An electroluminescent device as claimed in any one preceding claim wherein the non-polymeric material is selected from compounds of formula I-VI, wherein substituents R_1 - R_4 are independently selected carbyl-derived groups, including hydrocarbyl-derived groups, including alkyl, alkoxy, alkyleneoxy, thioalkyl, acyl, and aryl groups, all of which are optionally substituted, or other substituents including halo, hydroxy, amino, cyano or nitro.

SMC 60569

27



32. An electroluminescent device as claimed in any one of claims 1 to 28 wherein the non-polymeric material is an electron transporting material selected from AlQ3, bathophenanthroline, PBD, CuPc (copper phthalocyanine), PTCDA.

SMC 60569

28

33. An electroluminescent device as claimed in any one preceding claim wherein the composition is solution coated.
- 5 34. An electroluminescent device as claimed in any one preceding claim wherein the composition is applied by a coating technique selected from dip coating, roller coating, reverse roll coating, bar coating, spin coating, gravure coating, lithographic coating (including photolithographic processes), ink jet coating (including continuous and drop-on-demand, and fired by piezo or thermal processes), screen coating, spray coating and web coating.
- 10 35. An electroluminescent device as claimed in any one preceding claim wherein the layer comprising the composition has a thickness from 10 nm to 500 nm.
- 15 36. An electroluminescent device as claimed in claim 35 wherein the layer comprising the composition has a thickness from 40 nm to 500 nm.
- 20 37. An electroluminescent device as claimed in claim 36 wherein the layer comprising the composition has a thickness from 60 nm to 500 nm.
- 25 38. An electroluminescent device as claimed in claim 37 wherein the layer comprising the composition has a thickness from 100 nm to 500 nm.
- 30 39. An electroluminescent device as claimed in claim 38 wherein the layer comprising the composition has a thickness from 200 nm to 500 nm.
40. An electroluminescent device as claimed in any one of claims 35 to 39 wherein the thickness is up to 400 nm.
41. An electroluminescent device as claimed in any one of claims 35 to 40 wherein the thickness is up to 300 nm.
- 35 42. An electroluminescent device as claimed in any one preceding claim wherein the composition has a carrier (hole or electron) mobility greater than $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.
43. An electroluminescent device as claimed in any one preceding claim wherein the composition forms an ohmic interface between the composition and the electrode (anode or cathode).

SMC 60569

29

44. An electroluminescent device as claimed in any one preceding claim wherein the composition has a HOMO energy level in the range 4.8-5.2 eV.
- 5 45. An electroluminescent device as claimed in any one preceding claim wherein the layer comprising the composition is in direct contact with either the anode or cathode.
- 10 46. An electroluminescent device as claimed in any one preceding claim wherein the layer comprising the composition is the only layer present between the anode or cathode and the layer comprising the electroluminescent material.
47. An electroluminescent device as claimed in any one of the preceding claims wherein the composition is mixed in a binder resin.
- 15 48. An electroluminescent device as claimed in claim 47 wherein the binder resin is selected from the group: polyamide, polyurethane, polyether, polyester, epoxy resin, polyketone, polycarbonate, polysulphone, vinyl polymer, polystyrene, polyacrylamide, copolymers thereof and/or mixtures thereof.
- 20 49. An electroluminescent device as claimed in any one of the preceding claims wherein the layer comprising the composition is crosslinked.
50. An electroluminescent device as claimed in claim 49 wherein the layer is crosslinked by crosslinking of the polymeric material of the composition and/or
25 by crosslinking of the binder resin.
51. An electroluminescent device as claimed in any one of the preceding claims which is a top emission device wherein the transparent electrode is deposited after the layer comprising the composition.
30
52. A method of forming an electroluminescent device as claimed in any one of the preceding claims comprising depositing from a solution the layer comprising the composition.
- 35 53. A method of forming an electroluminescent device as claimed in claim 52 further comprising depositing at least one other layer by vapour deposition.
54. An electroluminescent device substantially as herein described.

SMC 60569

30

55. A method of forming an electroluminescent device substantially as herein described.

5

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.